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RADIATION EFFECTS IN SILICON SOLAR CELLS

QUARTERLY REPORT

by

R. A. Berger, H. Horiye, J. A. Naber, B. C. Passenheim

OCTOBER 10, 1969

PREPARED FOR

CALIFORNIA INSTITUTE OF TECHNOLOGY
Jet Propulsion Laboratory
4800 Oak Grove Drive
Pasadena, California 91103

CONTRACT 952387

This work was performed for the Jet Propulsion Laboratory, California Institute of Technology, as sponsored by the National Aeronautics and Space Administration under Contract NAS7-100.

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FOREWORD

This report contains information prepared by Gulf General Atomic Incorporated under Jet Propulsion Laboratory subcontract. Its contents are not necessarily endorsed by the Jet Propulsion Laboratory, California Institute of Technology, or the National Aeronautics and Space Administration.

ABSTRACT

The overall purpose of this program is to ascertain the nature of the defect or defects responsible for the degradation in output of silicon devices (solar cells) irradiated by space radiation. When the nature of the defects and their annealing mechanisms are known, it will be possible (1) to determine the parameters that will lead to the development of radiation-hardened devices and (2) to predict the effects of radiation and annealing on solar cells.

The use of electron spin resonance technique has been used to study the mechanisms for the production and annealing of the divacancy (Si-G7) in lithium-diffused n-type silicon irradiated with 30-MeV electrons. The results indicate that lithium does affect divacancy production and annealing in a manner similar to the manner in which lithium affects the production and annealing of the oxygen-vacancy (Si-B1) center.

Measurements of the lithium concentrations present in a lithium-diffused n-type silicon sample based on neutron activation analysis indicate that more lithium is present than as determined by resistivity measurements.

Studies to determine the nature of the recombination center in lithium-diffused silicon for the preirradiation, postirradiation-preanneal, and postanneal conditions are continuing.

CONTENTS

FIGURES

TABLES

1. INTRODUCTION

The overall purpose of this program is to ascertain the nature of the defect or defects responsible for the degradation in output of silicon devices (solar cells) irradiated by space radiation. When the nature of the defects and their annealing mechanisms are known, it will be possible (1) to determine the parameters that will lead to the development of radiation-hardened devices, and (2) to predict the effects of radiation and annealing on solar cells.

The present effort is concentrated on the study of the effects of lithium on the production and annealing of damage in silicon. This work is being performed on lithium-diffused bulk silicon using electrical measurements, such as minority-carrier lifetime, electron spin resonance (ESR), and electrical conductivity. The temperature range from 77.5° to 400°K is under investigation. The damage is introduced by 30 MeV electrons and fission neutrons.

During this reporting period, the electron spin resonance experiment received the most attention. Preparations were made for future minority carrier lifetime studies.

2. PROGRESS

2.1 MINORITY CARRIER LIFETIME

Studies of minority carrier lifetime have been aimed at determining the position of the recombination center in lithium-diffused silicon for preirradiation, postirradiation-preanneal, and postanneal conditions. Figure 1 shows the inverse temperature dependence of the lifetime from 130° to 380° K for a lithium-diffused silicon sample prior to irradiation.

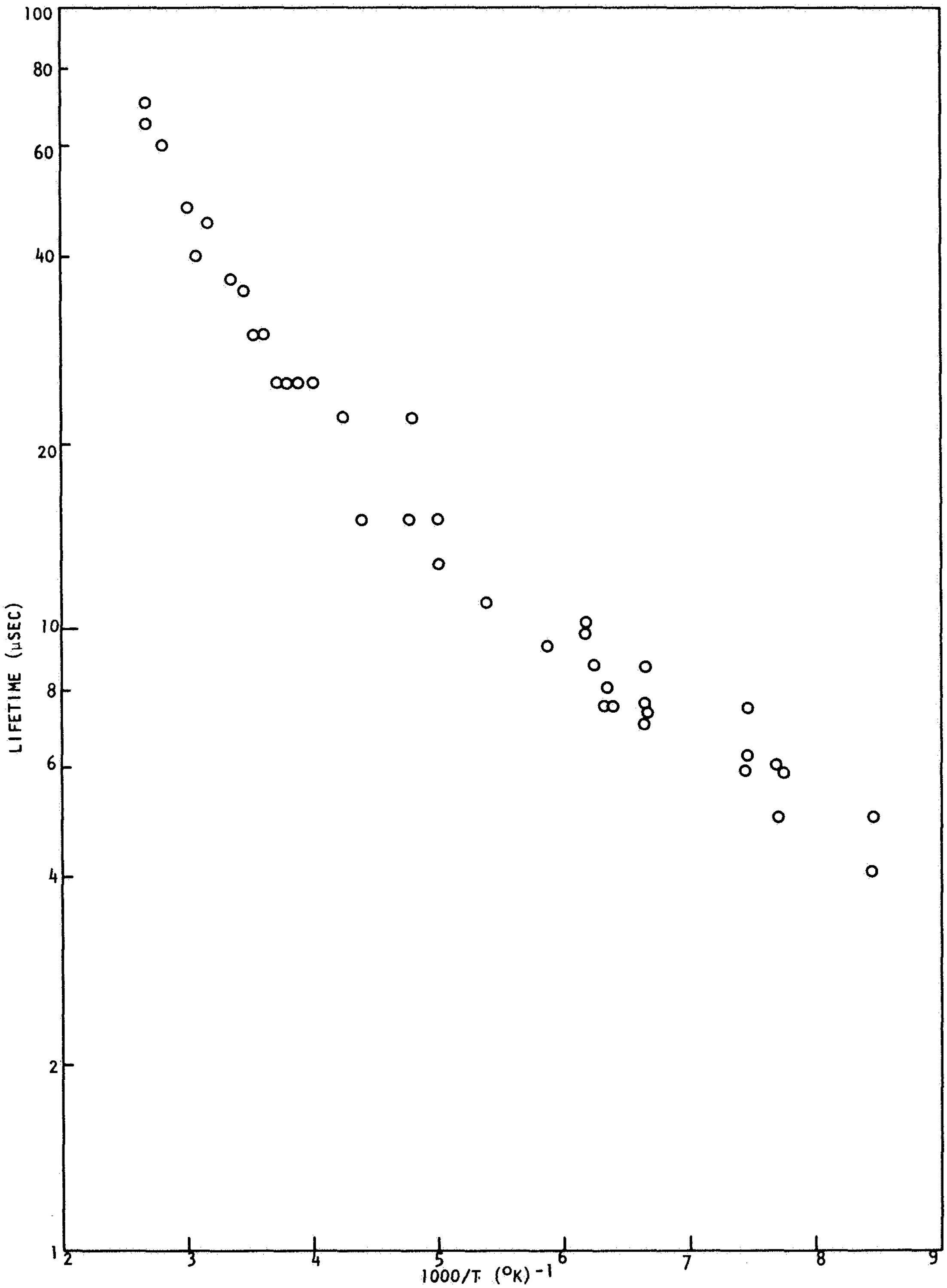

Fig. 1--Inverse temperature dependence of lifetime for lithium-diffused n-type silicon in preirradiation state

Figure 2 shows the inverse temperature dependence of the resistivity. To analyze this lifetime data properly, a number of temperature dependent factors must be considered.

The theory of recombination of excess carriers has been treated by others,[1-3] and the relation between theory and experimentally measured quantities has been reported earlier.[4-6] The conclusions can be summarized as follows. The Shockley-Read theory for a single-level defect assumes that the number of recombination centers is small relative to the excess-carrier density. This assumption implies that the excess electrons and holes have equal densities and lifetimes. The expression for the lifetime in this case is

$$\tau = \tau_{p_0}\left(\frac{n_0 + n_1 + \Delta n}{n_0 + p_0 + \Delta n}\right) + \tau_{n_0}\left(\frac{p_0 + p_1 + \Delta p}{n_0 + p_0 + \Delta n}\right) ,$$

where n_0 and p_0 are the thermal equilibrium electron and hole concentrations, n_1 and p_1 are the electron and hole concentrations calculated when the Fermi level is assumed to lie at the recombination center level, $\Delta n = \Delta p$ is the excess-carrier concentration, τ_{n_0} is the lifetime for electrons in highly p-type material, and τ_{p_0} is the lifetime for holes in highly n-type material. In n-type material (for p-type, the n's and p's are interchanged) where $n_0 \gg p_0$, dividing by n_0 gives

$$\tau = \frac{\tau_{p_0}\left(1 + \frac{n_1}{n_0}\right) + \tau_{n_0}\left(\frac{p_1}{n_0}\right) + \frac{\Delta n}{n_0}\left(\tau_{p_0} + \tau_{n_0}\right)}{1 + \frac{\Delta n}{n_0}}$$

or

$$\tau = \frac{\tau_\ell + \tau_h\left(\frac{\Delta n}{n_0}\right)}{1 + \frac{\Delta n}{n_0}} ,$$

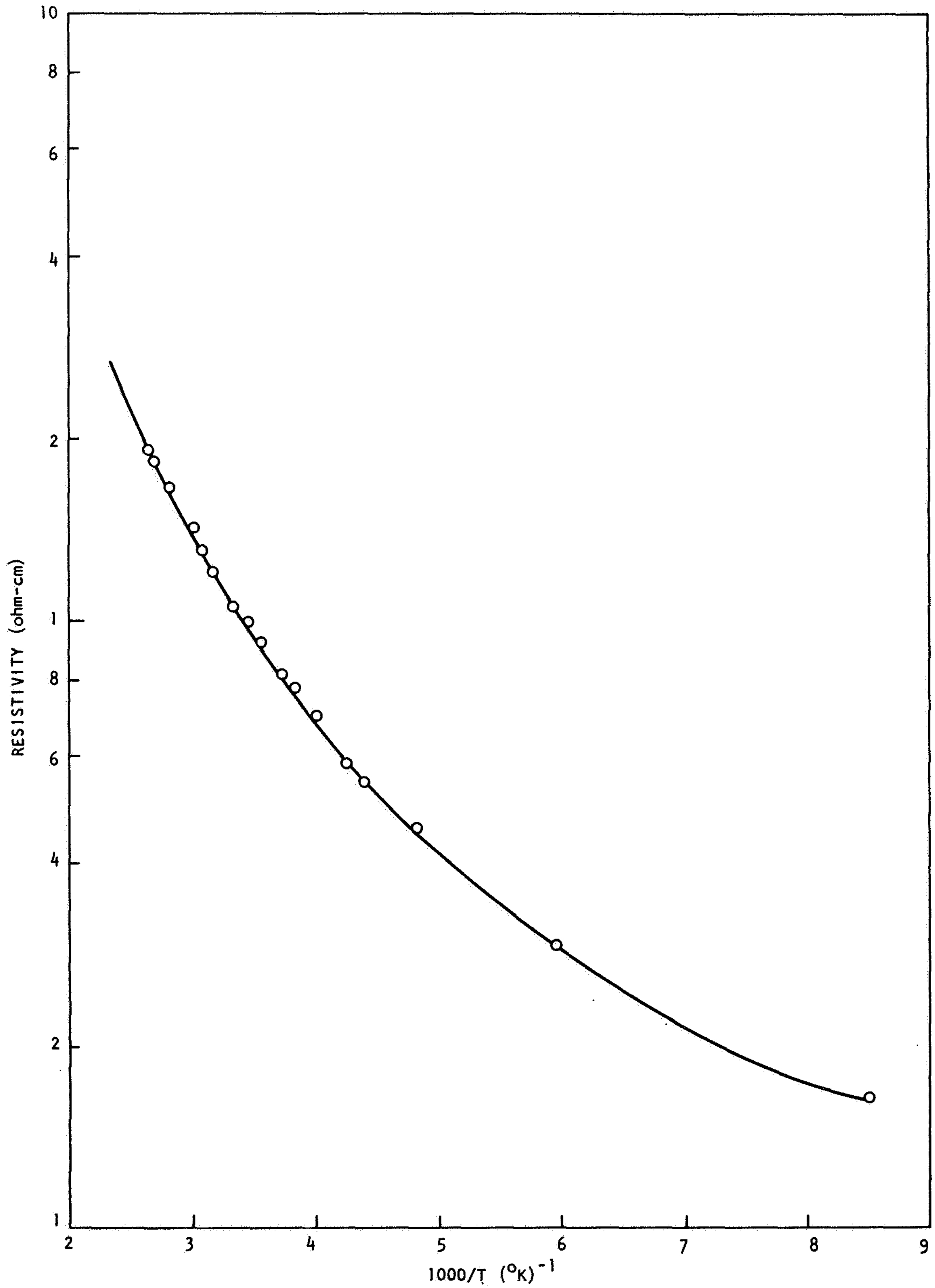

Fig. 2--Inverse temperature dependence of resistivity for lithium-diffused n-type silicon in preirradiation state

where τ_ℓ (the low-injection-level lifetime) and τ_h (the high-injection-level lifetime) are the corresponding terms in the equations. A plot of $\tau\ (1 + \Delta n/n_0)$ versus $\Delta n/n_0$ yields τ_ℓ as the intercept and τ_h as the slope. Linearity of this plot is a test of the validity of the theory.

The temperature dependence of τ is found in terms containing τ_{p_0}, τ_{n_0}, n_1, p_1, and n_0. The term n_0 is known, however, and can be extracted from the data. The temperature dependences of τ_{p_0} and τ_{n_0} are exhibited through the thermal velocity V_{th} of the carriers and the capture cross section σ, where

$$\tau_{p_0} = \frac{1}{N_R\ V_{th}\ \sigma_p}$$

and

$$\tau_{n_0} = \frac{1}{N_R\ V_{th}\ \sigma_n}$$

and where N_R is the concentration of recombination centers.

Theoretical work on the temperature dependence of the cross section for neutral and attractive recombination centers has been performed by Lax.[7] A brief summary of the theory is given in Ref. 8. Figure 3 gives the temperature dependence for the cross sections of the singly charged attractive center and the neutral center.

A great number of investigators do not consider the temperature dependence of all the terms in Eq. (1). At the present time, the data of Fig. 1 has not been successfully analyzed so that the position of the recombination center can be determined. The Fermi level during the measurement moved from 0.13 eV to 0.45 eV below the conduction band. If the recombination center is located in this range, proper analysis of the lifetime data will determine its position.

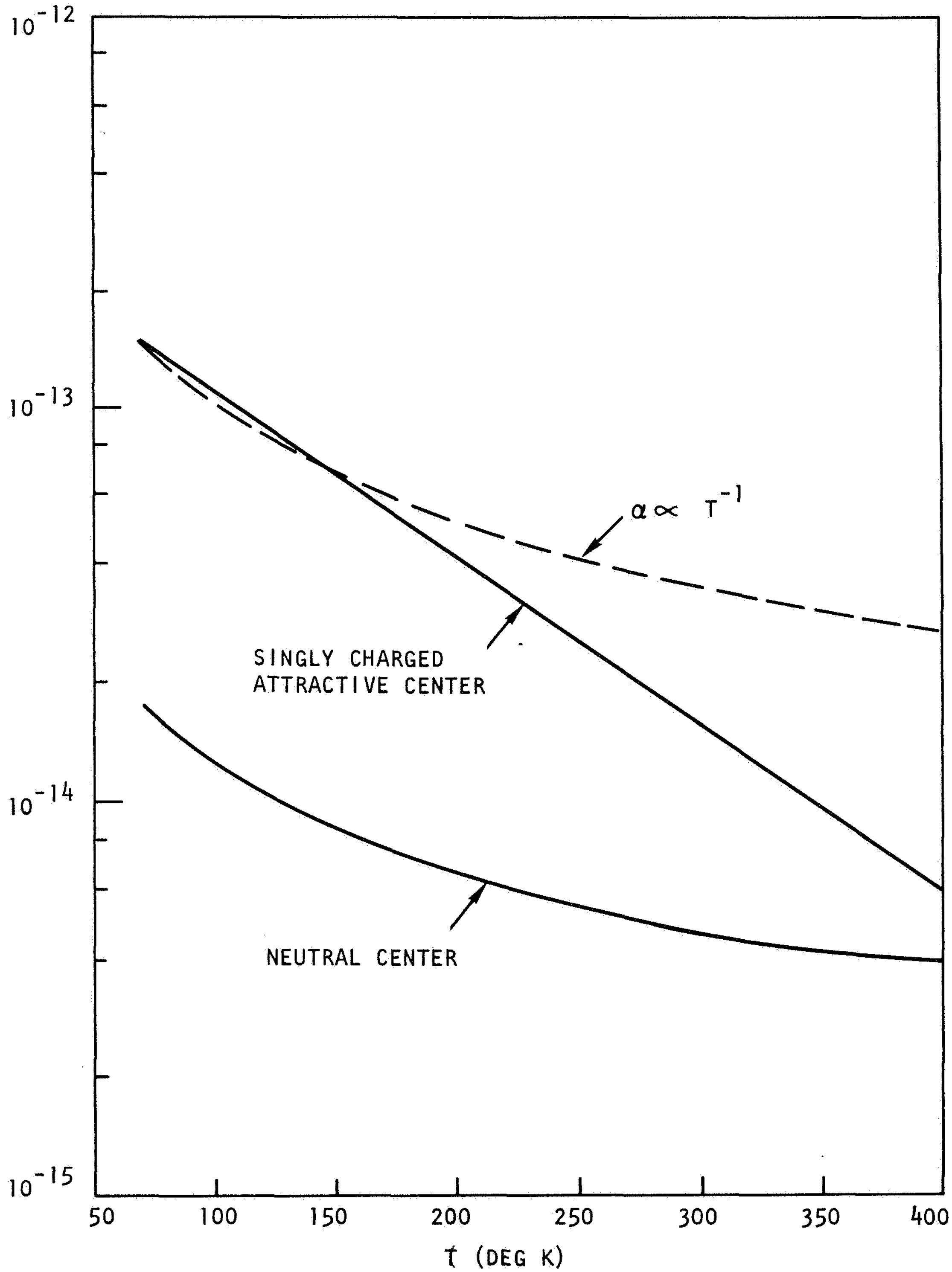

Fig. 3-- Recombination cross section in silicon versus temperature

The analysis of this data is being pursued at the present time. The experiments to determine the temperature dependence of the lifetime for samples in postirradiation-preanneal and postanneal conditions are being performed and will be analyzed to determine the position and nature of the recombination center in these conditions.

A paper entitled "Minority Carrier Lifetime Degradation and Anneal in Neutron-Irradiated Lithium-Diffused n-Type Silicon" has been prepared for publication in open literature. The paper has been approved for publication by Jet Propulsion Laboratory and has been submitted to Radiation Effects.

2.2 NEUTRON ACTIVATION ANALYSES

Low-resistivity lithium-diffused silicon samples were prepared by the lithium-oil paint-on technique.[9] These samples contained large quantities of lithium; resistivity measurements indicated lithium concentration of 10^{17} atoms/cm^3. The activation analysis measurements indicated lithium concentrations up to eight times greater than that indicated by the resistivity measurements.

The general trend of these activation analysis measurements leads us to believe that there is more lithium present in the lithium-diffused samples than indicated by resistivity measurements. This lithium may be present as a precipitate in the sample.

The possibility of freeing this precipitated lithium adds new possibilities to the annealing mechanism and methods of annealing present in lithium-diffused silicon.

2.3 ELECTRON SPIN RESONANCE (ESR)

Electron spin resonance has been a powerful technique in the study of radiation effects in silicon. ESR is one of the few techniques[10] which provides information about the detailed nature of the defects.

ESR techniques have been used quite successfully at Gulf General Atomic. Programs[11-14] investigating the production, annealing, and properties of various damage centers including the B1, G6, G7, and G8 have been completed. Recently, the ESR technique has been used to study the effect of lithium in radiation damage. A thorough investigation of lithium on the B-1 (oxygen-vacancy) center was completed during an earlier contract.[12] This study was of particular value since many investigators feel that the B-1 center is the predominant recombination center in silicon irradiated with 1-MeV electrons. The results of this study provided invaluable insight into the interaction of lithium with radiation-produced entities including impurity-related defects.

During the present contract, the ESR technique is to be used to gain more fundamental information on the role of lithium in displacement damage processes. The first study is the investigation of the effects of lithium on the production and annealing of the divacancy. The divacancy is an important damage center to study, for it is thought to be one of the recombination centers present in silicon after high energy electron and neutron radiation.

2.3.1 Theory

The divacancy is thought to have three electrical levels within the forbidden gap. These levels are located at 0.17 eV and 0.4 eV below the conduction band and at 0.25 eV above the valence band. If the Fermi level is (1) above 0.17 eV, the divacancy is in a double negative charge state and nonparamagnetic; (2) if between 0.17 and 0.4 eV, the divacancy is in a single negative charge state and paramagnetic; and (3) if below 0.4 eV, the divacancy is neutral and nonparamagnetic.(10,15) This is diagrammed in Fig. 4.

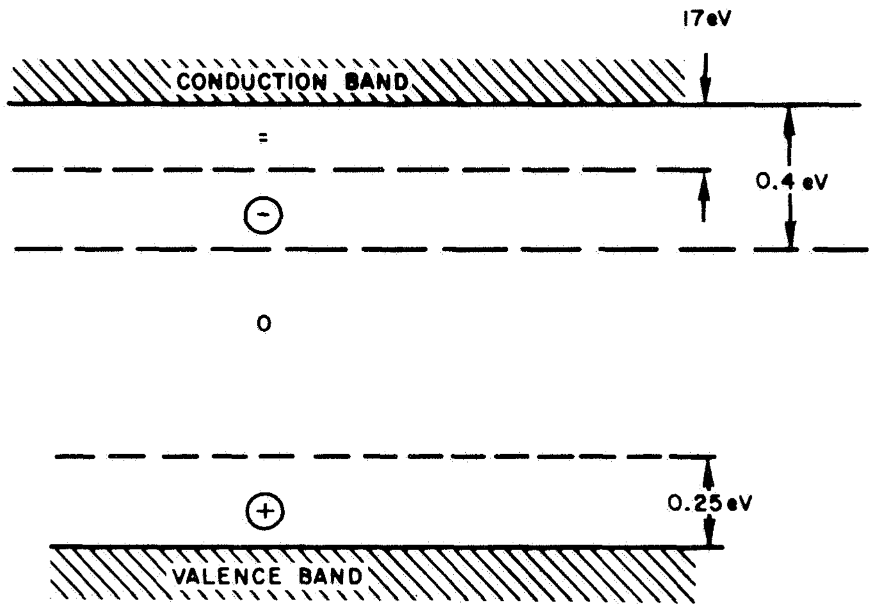

Fig. 4--Energy level diagram for the divacancy in silicon

For n-type silicon the level below the middle of the forbidden gap is always filled and is not observable by ESR techniques.

A plot of the g values of the paramagnetic divacancy as a function of the magnetic field direction is given in Fig. 5. The magnetic field is rotated in a (110) plane and θ is the angle between <100> direction in the crystal and the magnetic field. The principal values for the diagonalized divacancy g tensor (Si-G7) are:

$g_1 = 2.0012$

$g_2 = 2.0135$

$g_3 = 2.0150$

with $\phi = 29°$ from the [011] axis in the (011) plane.(10)

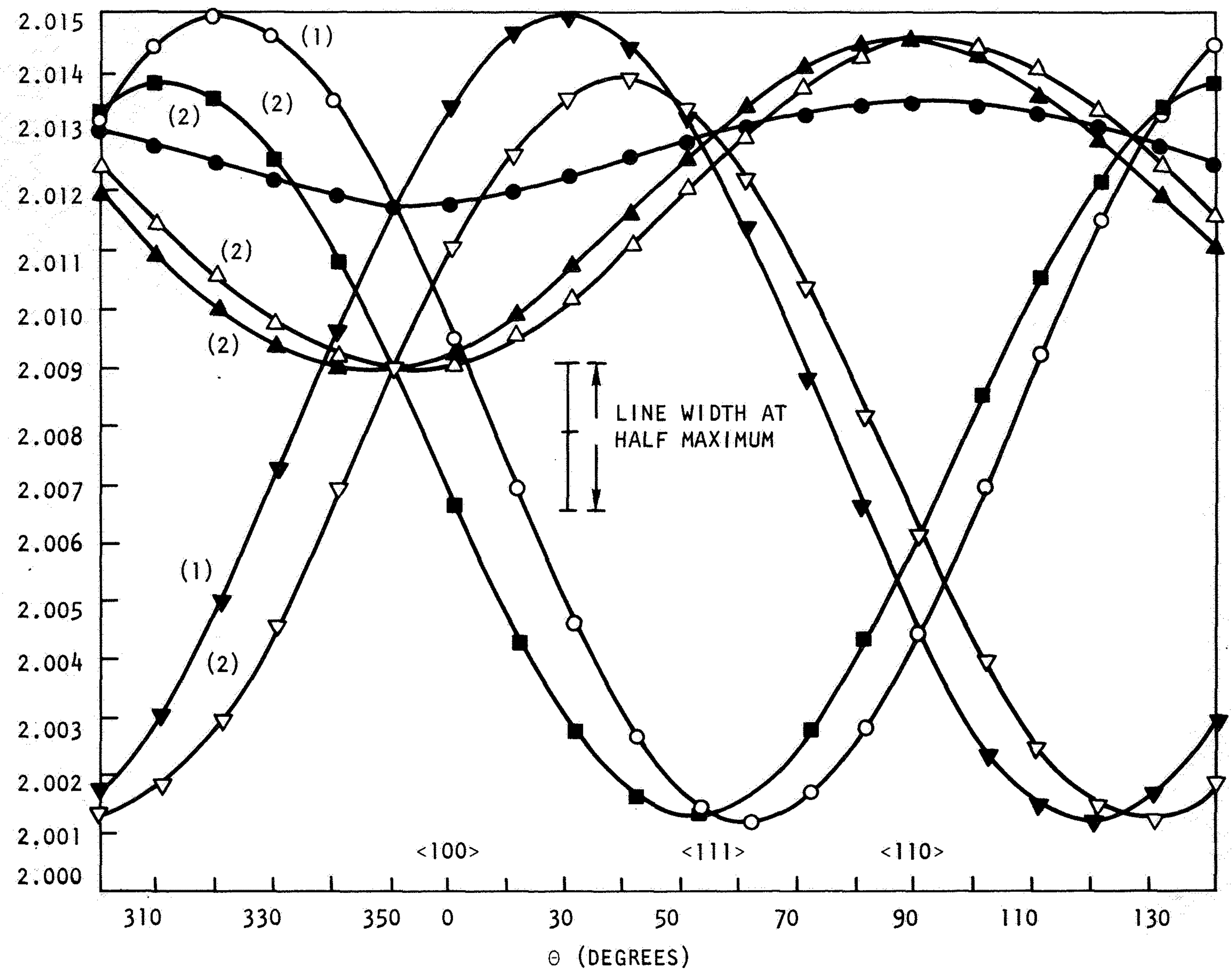

Fig. 5--g values of the paramagnetic divacancy as a function of the magnetic field direction

2.3.2 Experiment

Samples for the ESR studies were produced by the diffusion of lithium in 10^4 ohm-cm float zone n-type silicon. The lithium paint-on technique was used to diffuse lithium into the samples. The samples prepared ranged in resistivity from 0.11 to 0.66 ohm cm. This corresponds to a room temperature carrier concentration from 10^{17} to 10^{16} carriers/cm^3.

The first three samples of 0.66 ohm cm were irradiated with 30 MeV electrons to fluencies of 1×10^{16}, 2×10^{16}, and 3×10^{16} e/cm^2. Immediately after the room temperature irradiations, the samples were stored in liquid nitrogen.

The samples were briefly brought to room temperature for resistivity measurements using a four-probe apparatus. The room-temperature resistivities, carrier concentrations, and Fermi levels are presented in Table 1.

TABLE 1

ACCUMULATIVE ELECTRON FLUENCE FOR SAMPLES 1, 2, AND 3

Sample Number	Fluence (electrons/cm^2)	Postirradiation Resistivity (Ω-cm) 300°K	Carrier Concentrations (number/cm^3)	Fermi Level (eV below conduction band
1	1×10^{16}	2.9	1.5×10^{15}	0.26
2	2×10^{16}	35.	1.5×10^{14}	0.32
3	3×10^{16}	9900.	$\sim 10^{12}$	$\sim$ 0.45

The Fermi-level and conduction electron density was determined from the postirradiation room temperature resistivity. Previous[12] measurements yield an approximate divacancy introduction rate of

$$\frac{\Delta n}{\Delta \Phi} = 0.1 \ \frac{\text{divacancies}}{\text{e-cm}}$$

The divacancy density was estimated from the introduction rate and the measured fluence. The density of paramagnetic divacancies at 20°K was also estimated. Any annealing of the divacancy because of lithium migration due to the room temperature irradiation was neglected in the above estimates since it was thought to be insignificant.

The results of the estimates predicted that a 20°K Sample 1 should have had about 1×10^{15} divacancies/cm^3 in the double negative charge state, Sample 2 should have had 1.85×10^{15} divacancies/cm^3 in the single negative charge state, and Sample 3 should have had only 3.6×10^{14} divacancies/cm^3 in the single negative charge state. Thus, only Sample 2 contained enough paramagnetic divacancies to be seen in our ESR spectrometer since the noise level of our spectrometer(16) is 4×10^{14} spins/cm^3.

However, when Sample 2 was inserted into the ESR spectrometer and a careful search was made for the signal above the noise level of the spectrometer, no resonance peak was observed. Part of this search was concentrated at a magnetic field of 3312 gauss parallel to the <111> axis where a predicted(16) signal with a signal-to-noise ratio of 3.6 should have been observed. No resonances were observed in any of the three samples when they were searched over several magnetic field directions and over a range of 60 gauss.

The results of these experiments lead to two possible conclusions:

1. The divacancies had a lower introduction rate due to the effect of the presence of lithium.
2. There was significant annealing of the divacancy during the irradiation. This annealing was due to migration of lithium to the divacancies.

To eliminate the possibility of thermal annealing due to the migration of lithium to the divacancy, it was decided to perform the irradiations at 77°K and store the sample at this temperature. Three ESR samples (Samples 4, 5, and 6) were prepared from 10^4 ohm-cm float zone n-type silicon. After lithium diffusion each had a room temperature resistivity of 0.11 ohm-cm corresponding to a carrier concentration of 10^{17} cm^{-3}. The total defect introduction rate and divacancy introduction rate are different at 77°K than at 300°K, so to establish these rates it was decided to irradiate Samples 4, 5, and 6 in small fluence steps. Table 2 shows the fluence received by each sample.

TABLE 2

ACCUMULATIVE ELECTRON FLUENCE FOR SAMPLES 4, 5, AND 6

Sample No.	Fluence at End of Irradiation No. 1 (e/cm^2)	Fluence at End of Irradiation No. 2 (e/cm^2)	Fluence at End of Irradiation No. 3 (e/cm^2)
4	4×10^{16}	1.2×10^{17}	2.8×10^{17}
5	6×10^{16}	1.6×10^{17}	3.2×10^{17}
6	8×10^{16}	2.0×10^{17}	4.0×10^{17}

A preliminary estimate of the sample resistivity after irradiation can be made by observing the Q (quality factor) of the ESR spectrometer sample cavity at 77°K, with the sample in place. If the Q is low, the sample has not been sufficiently damaged--not enough electrons have been removed from the conduction band to put enough divacancies in the paramagnetic charge state. At the end of the irradiation No. 1, none of the three samples showed a reasonable Q at 77°K. At the end of irradiation No. 2, only Sample 6 had a good Q, and Sample 4 had a low Q. This establishes a total introduction rate for these samples at 77°K of about 0.5 $(e/cm)^{-1}$ since the Q of the sample cavity started to change at fluences of 2×10^{17} e/cm^2.

Samples 4, 5, and 6 were examined at 20°K in the ESR spectrometer following irradiations 2 and 3, but no paramagnetic resonances were detected. Either the introduction rate of the divacancies was too low, or the divacancies produced were in the wrong charge state, i.e., they were nonparamagnetic instead of paramagnetic.

For the purpose of determining the introduction rate of paramagnetic divacancies, consider Sample 6 at the end of irradiation 2 (total fluence of 2 x 10^{17} e/cm^2). The calculated noise level for this sample is 5 x 10^{14} spins/cm^3. Since no divacancy <111> axis signal was detected, the divacancy introduction rate must be less than 0.004 divacancies/e-cm. Otherwise, the <111> peak should have been detected. Previous data[13] on a 0.1 ohm-cm phosphorus-doped float zone silicon sample irradiated at 77°K with 30-MeV electrons yielded a divacancy introduction rate of 0.04 divacancies/e-cm, which is well above the apparent introduction rate of paramagnetic divacancies in lithium-diffused silicon. This implies that the divacancy introduction rate at 77°K is much lower for lithium-diffused silicon than in phosphorus-doped silicon, **if** all divacancies which are produced are paramagnetic.

Another possibility for not observing divacancies is that the introduction rate of divacancies may be the same for lithium-diffused silicon as for nonlithium-diffused silicon (0.04 divacancies/e-cm) at 77°K, but that the divacancies were not detected in the ESR spectrometer because they were in a nonparamagnetic charge state. As previously mentioned, observation of the Q as a function of fluence indicated a carrier removal rate of 0.5 cm^{-1} or a fluence of 2 x 10^{17} e/cm^2 was required to substantially depopulate the conduction band, but no divacancies were observed up to this fluence. After irradiation to a fluence of 2.8 x 10^{17} e/cm^2, Sample 4 was examined at 0°C with a two-probe resistivity measuring device and was found to have less than 10^{12} carriers/cm^3. This means the Fermi level is below 0.4 eV so that nearly all the divacancies in this sample were in a neutral charge state and, therefore, not paramagnetic at the 20°K ESR measurement temperature. Thus, for these samples, the divacancy is in the paramagnetic charge state for some fluence between 1.2 x 10^{17} and 2.8 x 10^{17} e/cm^2. Furthermore, the limited range of fluences (or window) over which the divacancies are paramagnetic must be less than the 4 x 10^{16} e/cm^2 fluence increments between the samples for irradiation 2 or 8 x 10^{16} e/cm^2 between irradiation 2, Sample 6 and irradiation 3, Sample 4. An estimate of the probable width of the "window" is about 1.1 x 10^{16} e/cm^2 for Samples 4, 5, and 6. With a window width of only about 0.11 x 10^{17} e/cm^2

for a fluence of 2×10^{17} e/cm^2, it is easy to miss the "window" on irradiations 2 and 3 for Samples 4, 5, and 6.

The temperature of Samples 4, 5, or 6 was not raised above 77°K since the elimination of the possibility of migration of lithium to the divacancies was important.

After irradiation 3, Sample 6 was illuminated with an incandescent light for three hours while in the spectrometer cryostat at 20°K in an attempt to change the charge state of the divacancies from nonparamagnetic to paramagnetic. At temperatures near 20°K the time constants for returning to equilibrium can be very long;[(13)] therefore, populated divacancies (paramagnetic) would exist for long periods of time. If this were true, then the ESR resonances of these paramagnetic centers would be seen. However, in the above experiment on Sample 6, no new resonance was observed after the illumination.

To make the ESR measurement when the divacancies are in the paramagnetic charge state for irradiation at 77°K would require continuous resistivity measurements on a sample of ever increasing resistivity. The irradiations would be done at 77°K and the resistivity measurements would be made at a higher temperature, probably 0°C, for convenience. Once all the divacancies were put into the paramagnetic charge state, no true annealing experiments could successfully be made, since other experiments[(17,18)] indicate an increase in resistivity upon annealing which lowers the Fermi level. This would make the divacancies nonparamagnetic and reduce the ESR signal. When this happens, it would not be possible to determine whether the divacancies disappeared due to the migration of lithium or due to depopulation of vacancies by Fermi level motion.

2.3.3 Conclusions

The study of the divacancy (Si-G7) in lithium-diffused silicon indicates for room temperature (300°K) irradiations that the divacancy is affected by the presence of lithium. The overall effect of lithium is to decrease the number of divacancies present after a 300°K irradiation. The lithium either lowers the introduction rate of divacancies or anneals the divacancies once they are formed. Earlier work[18] has shown that both of these mechanisms are present in B-1 center production and annealing.

The ESR measurements of the divacancy were performed after irradiations at 77°K in the hope of separating the mechanisms responsible for the decrease in the number of divacancies observed after room temperature irradiations of lithium-diffused n-type silicon. These experiments were inconclusive for the reasons presented in Section 2.3.2.

The study of the Si-G7 and B-1 centers has shown that lithium is effective in decreasing the number of these centers produced in 300°K electron irradiations. Since both of these centers are considered as recombination centers in electron irradiated nonlithium-diffused silicon, there is a possibility of decreasing the lifetime degradation in silicon due to the presence of lithium. However, experimental measurements[18] of the lifetime degradation constant for 30-MeV electrons at 300°K yields a higher degradation rate for the lifetime in lithium-diffused silicon than in nonlithium-diffused silicon. This implies that in electron irradiations of lithium-diffused silicon a new recombination center is produced which degrades the lifetime faster than either the divacancy or B-1 center. It is most logical that this new recombination center contains lithium or is affected in its production by lithium.

To make the above argument more complete, other centers that are thought to be recombination centers must be studied in lithium-diffused silicon. The above results will now be supplemented by ESR studies of the Si-G8 center (vacancy-phosphorus pair) in lithium-diffused silicon. The Si-G8 center is better suited to study by ESR because it is paramagnetic over a much wider range of fluences than the divacancy. The 30-MeV electron irradiations will be performed at 77°K to prevent migration of the lithium. This experiment will give us information on the introduction rate and annealing of the Si-G8 center in lithium-diffused phosphorous doped silicon. The study of the divacancy and B-1 center gave us information on the annealing of a negative charged defect by lithium. The paramagnetic state of the Si-G8 is a neutral defect, so its annealing by lithium should be different from the annealing of the divacancy and B-1 center by lithium.

It is anticipated that by comparison of various specific defect introduction rates that the relative cross sections for production of various vacancy-impurity defects can be determined. The study of the annealing rates by lithium of various vacancy-impurity defects will supply information on the charge-state dependence of the defect annealing.

At the conclusion of these systematic studies, it would be possible to select or at least designate the characteristics of the proper dopants (impurities) for silicon which is to be lithium diffused, so that the recombination centers formed by irradiation have a minimal effect on lifetime degradation. The lithium will still be active in annealing the defects due to its migration. This approach will permit the determination of the parameters for silicon, used in solar cells, that will optimize their radiation resistance.

2.4 PLANS FOR THE NEXT REPORTING PERIOD

During the next reporting period, we plan to:

1. Determine lifetime degradation constant of lithium-diffused silicon as a function of temperature.
2. Determine effect of lithium depletion after electron and neutron irradiations on the steady-state photoconductivity signal.
3. Study the Si-G8 center using ESR.
4. Continue investigation of lithium content by neutron activation analysis.

2.5 NEW TECHNOLOGY

No new technology is currently being developed or employed in this program.

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